## **Supporting Information**

## Charge Transfer Induced Multifunctional Transitions with Sensitive Pressure Manipulation in a Metal-Organic Framework

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To further confirm the charge-transfer induced ferroelectric phase transition in the present Fe<sub>2</sub>Co-MOF, the ferroelectric hysteresis loops were measured following the electric polarization measurements. Figure S1 shows a typical P-E loop obtained at 10 K based on the "positive-up-negative-down (PUND)" method, which can exclude most extrinsic effects on ferroelectric polarization. Obviously, the polarization is switchable completely, revealing the occurrence of ferroelectric phase transition originated from the Fe-Co intermetallic charge transfer. However, we would like to point out that in an Fe<sub>2</sub>Co-MOF chemical formula, there exists four H<sub>2</sub>O, which is very easy to release in vacuum for measurement. Once the crystal water is released, the charge transfer will vanish and then no polarization taking place. During the measurements, we use some glues to cover the sample. Even for this protecting, some crystal water is still released. This is the reason why the polarization value obtained in P-E loop is different from that in P-T curve as represented in Fig. 4b. Anyway, the electric polarization is switchable completely in Fe<sub>2</sub>Co-MOF, and the charge-transfer induced ferroelectricity is intrinsic.



Figure S1. Ferroelectric hysteresis loop of Fe<sub>2</sub>Co-MOF measured at 10 K.